

Ultraviolet Photon Emission Observed in the Search for the Decay of the ^{229}Th Isomer

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Ultraviolet spectroscopy of ^{233}U and ^{232}U , in the form of uranium oxide dissolved in 3M nitric acid, has revealed a number of spectral lines with widths of less than 6 nm from the ^{233}U source but no detectable narrow lines from the ^{232}U source. These lines could possibly be attributed to the decay of the ^{229}Th isomeric state at an energy of around 4.0 eV. [S0031-9007(98)05793-7]

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In 1994 Helmer and Reich [1] determined the separation of the ground and first excited nuclear states in ^{229}Th to be 3.5 ± 1.0 eV. Recently the direct decay from this state may have been observed at higher resolution [2]. This is then the lowest excitation energy for a nuclear first excited state yet found and the first to be on a similar energy scale to atomic valence electrons. Such a small energy separation provides the potential to examine a regime of nucleus-atom interactions not previously observable. The value for the excitation energy was obtained by applying the Ritz combination principle to groups of γ -ray energies of several tens to hundreds of keV. Using energies of 4–5 orders of magnitude larger than the energy under investigation makes it difficult to increase the accuracy of the measured energy separation by this method.

The spins and parities of the ground and first excited states together with the small excitation energy suggest that the excited state should be a long lived isomeric state [3]. A photon emitted by the direct decay of the first excited state to the ground state would have a wavelength in the blue-ultraviolet region of the electromagnetic spectrum, i.e., between 270 and 500 nm, with 3.5 eV corresponding to 350 nm. Thus conventional UV spectroscopy is an ideal tool for increasing the accuracy to which the separation is known. A recent experiment [2] used UV spectroscopy of two solid sources of ^{233}U to look for the ^{229}Th -isomer deexcitation following the α decay of the ^{233}U . The results show a broad spectral feature at ~ 2.5 eV, with a set of spectral lines centered at approximately 3.5 eV.

In the current work a ^{233}U source was also used; however, the radioactive isotope was in the form of uranium oxide dissolved in 3M nitric acid, producing an equilibrium in which uranyl nitrate ions dominate (UO_2NO_3^+). The source activity was 4 MBq and its volume of 0.25 ml was contained in a 6 mm internal diameter quartz tube. ^{233}U has an α -decay half-life of 1.7×10^5 years, which ensures the activity will be essentially constant throughout the course of the experiment. It has been estimated that about (1–2)% of the total α decays of the uranium feed into the ^{229}Th first excited state [4,5]. Because both ^{233}U and ^{229}Th have

long half-lives it was not considered necessary to remove the daughters produced since the production of the ^{233}U source in 1992.

A control source, with ^{232}U as the main active element, was also made. The half life of ^{232}U is 72 yr, which, while adequate for the duration of the measurements means that a substantially smaller quantity of the isotope is needed to obtain the same activity as that of the ^{233}U source. To ensure the same chemical composition with the same uranium α activity, the uranium content of the ^{232}U source was matched to that of the ^{233}U source by adding depleted uranium oxide to the sample. The buildup of daughter products in the control source meant that the total α -particle activity was 8 MBq, with 4.5 MBq being due to the ^{232}U (ideally ^{234}U should be used, as a control, due to its similar half-life to ^{233}U , i.e. 2.5×10^5 yr, and the similarity of the α -particle and γ -ray energies). Both sources were produced by AEA Technology [6]. The control source was used to enable the fluorescence background contribution to be subtracted. More importantly, the absence of discrete lines in the ^{232}U spectrum gives added confidence that any discrete line structure in the ^{233}U spectrum could be attributed to the isomer decay and not some other process. The use of a control source and the carefully controlled chemistry of the radioactive sources used in this investigation represent a significant advance in experimental technique over previous investigations of the ^{229}Th isomer.

The UV spectrometer was a Jobin-Yvon imaging spectrograph (CP-140-1604) [7], of f number = 2. This has a concave diffraction grating blazed for 250 nm as the dispersing element. The spectral range covered by the spectrometer is 190 to 670 nm, with an optimum dispersion of 17 nm/mm at the focal plane. The detector placed at the focal plane of the spectrometer was an imaging photon detector (IPD), made by Instrument Technology Limited, with a MgF_2 entrance window. The detector itself consists of a S20 photocathode placed several centimeters behind the entrance window. The cathode diameter is 18 mm, giving a limit of 300 nm on the spectral range for a single position of the detector. The photocathode is followed by a stack of three microchannel plates (MCP's). Close

behind the final MCP is a position sensitive anode, with four arc-terminating resistors attached to the edges. Each of the four terminals of the anode has a fast preamplifier, which amplifies a fraction of the total pulse produced by the electrons from the MCP's hitting the anode. These four pulses are then shaped, buffered and fed into a separate processing unit to provide XY position information. An electromechanical shutter was placed intermittently between the source and the entrance slit of the spectrometer. For every 20 min of data taken with the source visible by the slit, another 20 min were taken with the source obscured. This allowed a systematic background subtraction to be made. A low-pressure spectroscopic mercury lamp was used for wavelength calibration of the spectrometer. In the current work an 8 mm high entrance slit of a width of 0.25 mm was used. This gave a resolution of 6.2 nm for the mercury lines. As the spectrometer has a 1:1 focusing arrangement the slit width imposes a limit of 4.2 nm on the resolution. A 0.1 mm slit produced a resolution of 5.0 nm but with a significant drop in count rate.

A similar experimental method was used by Irwin and Kim [2], as mentioned above. There were, however, some obvious differences with the work presented here. The two sources that they used were solid compounds of ^{233}U , one of which was uranyl oxide, whereas our sample was liquid with mainly uranyl nitrate ions. They used a monochromator, which allowed data to be collected in series, whereas our arrangement used a spectrometer and an IPD to collect the data in parallel.

The use of a liquid source provides some difficulties not encountered with a solid source. However, the fluorescence at around 360 nm excited by the α and γ activity, is broadened in the liquid and after background subtraction allows narrow features to be distinguished easily. This is, of course, one reason why a control source is required. A liquid source has other positive advantages in that the chemistry and physical structure is well defined. Furthermore, one does not have to worry about progressive lattice damage due to radioactive decay in a solid crystal source. Figure 1(a) shows the background subtracted results from a total of 14 days worth of running with the ^{233}U source. Immediately, it can be noted that there are two broad peaks, one at around 360 nm, and the other at 500 nm. The spectrum taken with the ^{232}U control source, Fig. 1(b), also exhibits a broad peak at 360 nm, which we ascribe to fluorescence in the liquid induced by radioactive decay. The intensity of γ radiation from the ^{232}U was also significantly higher than that from the ^{233}U source. This, together with the difference in α activity, makes it difficult to estimate the relative strength of the fluorescence from the two sources. Thus a scaling parameter was applied to the ^{232}U spectrum and this scaled spectrum subtracted from the ^{233}U data. The resulting spectrum reveals the details shown in Fig. 2 where the spectrum is plotted as a function of energy to facilitate a comparison with the results of Irwin and Kim [2]. It can be seen that as well as the broad

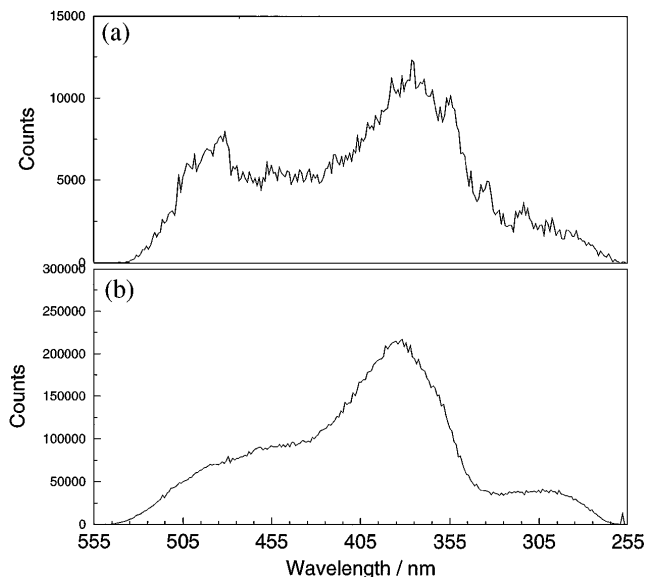


FIG. 1. (a) Upper plot: wavelength spectrum from the ^{233}U source after subtracting the counts obtained with the shutter closed. (b) Lower plot: spectrum for ^{232}U after background subtraction.

peak at around 2.5 eV, there are also 3 peaks, at 3.50 ± 0.04 , 3.71 ± 0.05 , and 3.96 ± 0.09 eV. The uncertainties quoted include calibration uncertainties. The widths of these latter peaks correspond within the uncertainties to the measurement of the widths obtained from the Hg calibration lines, and indicate a resolution of 6.2 nm.

Comparison between the data given in [2] and Fig. 1 shows that the broad and 3 narrower peaks appear at the same energies in the two experiments, suggesting that their production is not significantly influenced by the chemical surroundings. The fact that the spectra are similar, within the resolution of our experiment, may be accounted for by the way in which the Th ions in solution are surrounded by oxygen atoms bound in water molecules and nitrate ions. The coordination being similar in both the solid hydrated nitrate and the nitrate solution [8], one might expect similar chemical surroundings in the solid and liquid leading to similar optical spectra.

The presence of structure in the well-resolved lines in the ^{233}U source is somewhat of a mystery, although several possible explanations can be put forward. While it is clear that the structure seen in Fig. 2 arises unambiguously from the decay scheme of ^{233}U , it cannot automatically be attributed to the decay of the ^{229}Th isomer level. There are other differences between the two samples that might conceivably lead to the production of spectral lines in the ^{233}U sample and not in the control sample. For example, the decay chains of the two sources vary slightly. The decay of ^{233}U leads to actinium and francium isotopes while these elements are absent from the daughters

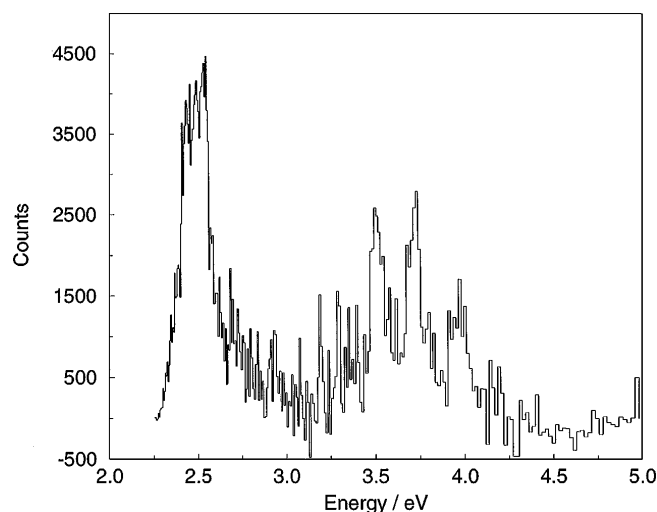


FIG. 2. Energy spectrum obtained by subtracting the scaled ^{232}U spectrum from the ^{233}U spectrum.

of the ^{232}U control source. The observed optical lines could be produced if the atomic electrons in the Ac (or Fr, its daughter product) were being deexcited and producing a fluorescence. The rate of detection expected for one fluorescence photon per Ac atom together with the 4 MBq activity of the source summed over 14 days (collecting time for the ^{233}U spectrum) gives a total number of detected photons commensurate with the total number of events in one of the narrower peaks. However, the set of spectral lines cannot at the present time be tied to a particular optical spectrum of any of the elements in the decay chain of ^{233}U . One might expect to see a variety of lines that could be attributed to a particular element if this were the mechanism producing the well-resolved structure. As no such assignments can be made we cannot make a definitive statement about the origin of the observed structures in the spectrum.

If the well-resolved structure does arise from the decay of the isomeric state, then this clearly does not have its origins in the hyperfine interaction, as this would require unfeasibly large magnetic fields. Irwin and Kim [2] suggest that the longer wavelength broad line (which from our work could conceivably be a triplet of narrow lines) is a result of the isomer decaying via an inelastic electronic bridge [9], where the nuclear isomer state decays through an interaction with the surrounding atomic electrons. It is worth noting that the isomer cannot decay via internal conversion because the ionization potential of the atom is greater than the excitation energy. It is possible that in fact the well-resolved structure starting at 3.5 eV also arises as a result of an interaction with

the atomic electrons and the isomeric energy is either the highest energy peak in the triplet (3.96 eV), or in fact at a yet higher energy >5 eV. Such an energy would not be observed in our work but would be within two standard deviations of the energy quoted by Helmer and Reich [1]. Clearly, such an approach would imply that the isomeric energy is higher than previous assessments of 3.5 eV and we might tentatively suggest that the excitation is more likely to be 4.0 eV. Other mechanisms such as a decay coupled to an atomic $M1$ transition, or a coupling to molecular vibrational levels could also conceivably give rise to a structured decay scheme.

As yet there is no direct evidence to confirm that any of these peaks are due unambiguously to the isomeric level. However, there are experiments that could be performed to aid our understanding of the origin of the observed spectral features. For example, a ^{229}Th sample in which the isomer has decayed would provide an excellent control source and would reveal whether the Ac and Fr daughters contribute narrow spectral lines to the fluorescence.

In summary, these results confirm, and increase the resolution of the peaks seen in [2]. Although seen in the energy region that would be expected by the direct deexcitation of the ^{229}Th first excited state, we urge caution in the assigning of the observed spectral features as arising from the decay of the nuclear isomeric state. Further work needs to be carried out to ascertain the cause of the observed structure.

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